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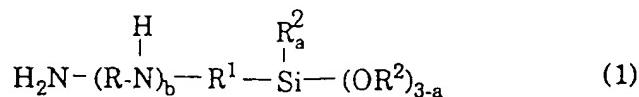
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(54) Primer composition

(57) A primer composition comprises (A) an organosilicon compound of the general formula (1)



wherein R is C<sub>1</sub>-C<sub>6</sub> alkylene, R<sup>1</sup> is a divalent hydrocarbon group having at least 4 carbon atoms, each R<sup>2</sup>, which may be the same or different, is an unsubstituted or substituted monovalent hydrocarbon group, a is 0 or 1 and b is 0, 1, 2 or 3, (B) a condensation catalyst for the organosilicon compound, and, optionally, (C) an organopolysiloxane resin.

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**Description**

This invention relates to a primer composition which ensures good adhesion between an adherend formed with an electrodeposition coating on the surface thereof and a room temperature curable silicone rubber body and is thus useful as a primer for sealants.

Silicone rubbers exhibit good heat resistance, cold resistance, weatherability and electric insulating properties, and have wide utility in the fields of sealing materials for buildings, adhesive sealing materials, electric insulating materials, potting materials and the like. Where strong adhesion is necessary between adherends and silicone rubbers as with sealants for buildings and adhesive sealing materials, usual practice is to pretreat the adherend with primers.

The primers employed for room temperature curable silicone rubbers which are used as a sealant for buildings include, for example, compositions which comprise silane coupling agents and/or reaction products thereof, polyester resin and acrylic resins, and compositions which comprise phenyl group-containing siloxanes, alkyl silicates and chlorosilanes.

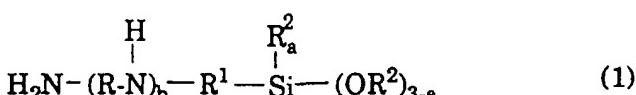
In recent years, acrylic resin paints or fluorine resin paints, which exhibit good weatherability, chemical resistance, electric insulating properties and inflammability, have been applied by electropainting or spraying and widely used as a coating for outer walls of buildings. Accordingly, there has been a demand of development of primers which have good adhesion to the coating films, especially to the films obtained by the electropainting.

Japanese Laid-open Application No. 2-219885 proposes a primer composition which comprises a hydroxyl group or alkoxy group-containing organopolysiloxane, an amino group-containing silane and a titanic acid ester. Japanese Laid-open Application No. 6-128553 proposes a primer composition which comprises a hydroxyl group or alkoxy group-containing organopolysiloxane, an organosilicon compound having an amino group and at least two alkoxy groups, an organosilicon compound having a mercapto group and at least two alkoxy groups, and an organotitanium and/or organozirconium compound. Although adhesion to metals to be painted is improved to some extent, it is not always satisfactory. Especially, the adhesion between an adherend such as an electropainted metal and a room temperature curable silicone rubber is not satisfactory.

It is accordingly an object of the invention to provide a primer composition which can overcome the problems of the prior art primer compositions and which is able not only to strongly adhere to adherends such as metals, glass, concrete and the like, but also to ensure strong bonding between adherends on which acrylic resins or fluorine resins have been electropainted and silicone rubbers.

The above object can be achieved, according to the invention, by a primer composition which comprises:

(A) an organosilicon compound of the general formula (1)



wherein R represents an alkylene group having from 1 to 6 carbon atoms, R<sup>1</sup> represents a divalent hydrocarbon group having not less than 4 carbon atoms, R<sup>2</sup>'s may be the same or different and independently represent an unsubstituted or substituted monovalent hydrocarbon group, a is 0 or 1, and b is 0, 1, 2 or 3; and

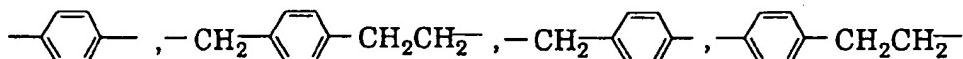
(B) a catalyst for condensation of the organosilicon compound.

(A) Organosilicon compounds

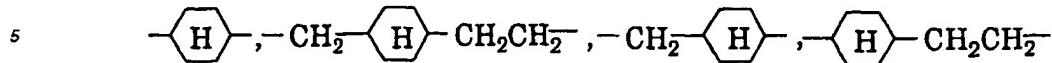
The organosilicon compounds used in the present invention are represented by the formula (1) and are an important component which ensures good adhesion imparted to the composition.

In the formula (1), R represents an alkylene group having from 1 to 6 carbon atoms. Specific examples include -CH<sub>2</sub>- , -CH<sub>2</sub>CH<sub>2</sub>- , -CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>- , -CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>- and the like, of which -CH<sub>2</sub>CH<sub>2</sub>- is preferred.

R<sup>1</sup> represents a divalent hydrocarbon group having not less than 4 carbon atoms, preferably 4 to 30 carbon atoms, and more preferably from 6 to 12 carbon atoms. If the number of carbon atoms is less than 4, satisfactory adhesion between an electropainted adherend and a silicone rubber may not be attained. Examples of the divalent hydrocarbon group include phenylene group-containing divalent hydrocarbon groups such as



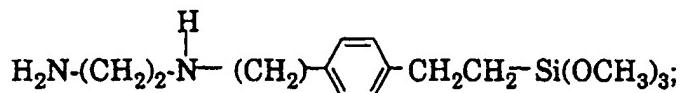
and the like, and cyclohexylene group-containing divalent hydrocarbon groups such as



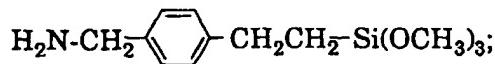
and the like, linear alkylene groups of the formula, -(CH<sub>2</sub>)<sub>n</sub>- wherein n is an integer of 4 or above such as -(CH<sub>2</sub>)<sub>4</sub>-, -(CH<sub>2</sub>)<sub>8</sub>-, -(CH<sub>2</sub>)<sub>12</sub>- and the like, and unsaturation bond-containing divalent hydrocarbon groups such as -CH=CH-CH=CH-. Of these, the phenylene group and the phenylene-group containing divalent hydrocarbon groups are preferred. If b is 0 or 1, R<sup>2</sup>s may be the same or different and represent a substituted or unsubstituted monovalent hydrocarbon group, preferably an alkyl group having from 1 to 4 carbon atoms, and more preferably methyl or ethyl. a is 0 or 1, preferably 0. b is 0 or an integer of 1 to 3, preferably 0 or 1.

Specific examples of the (A) component are those shown below:

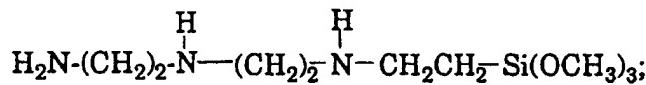
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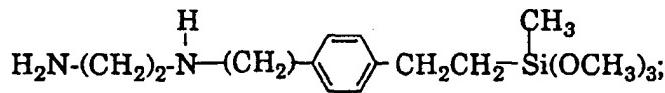


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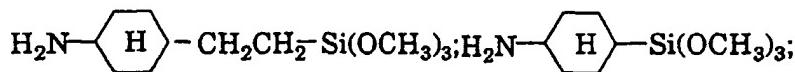


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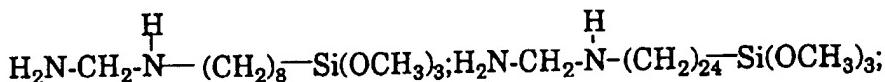
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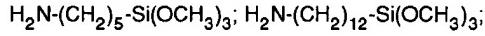
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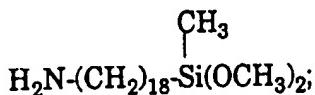


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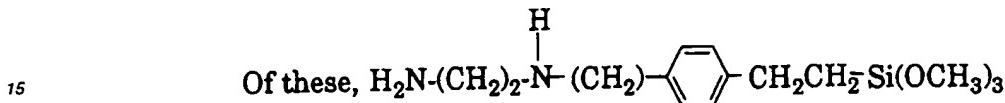


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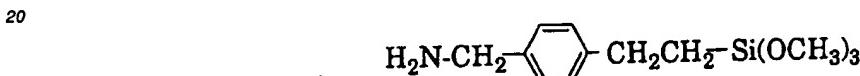




and



and



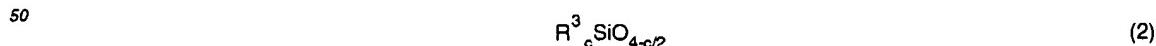
are preferred.

25 The condensation catalyst of the (B) component serves as a catalyst for curing the organosilicon compound of the (A) component. This component itself is taken in and becomes part of a final primer layer, thereby forming a strong film. The condensation catalysts include tin compounds, titanium compounds, zirconium compounds and aluminum compounds. Examples of the catalyst include titanium tetraalkoxides of the formula,  $\text{Ti}(\text{OR}^4)_4$  and/or partial hydrolyzates thereof, zirconium tetraalkoxides of the formula,  $\text{Zr}(\text{OR}^4)_4$  and/or partial hydrolyzates thereof, tin tetraalkoxides of the formula,  $\text{Sn}(\text{OR}^4)_4$  and/or partial hydrolyzates thereof, and aluminum trialkoxides of the formula,  $\text{Al}(\text{OR}^4)_3$  and/or partial hydrolyzates thereof, in which  $\text{R}^4$  is as defined with respect to  $\text{R}^2$  and preferably represents a monovalent hydrocarbon group having from 1 to 4 carbon atoms.

30 Specific examples of the titanium alkoxides include titanium tetramethoxide, titanium tetraethoxide, titanium tetrapropoxide, titanium tetraisopropoxide, titanium tetrabutoxide and the like, of which titanium tetraethoxide is preferred. Examples of the zirconium tetraalkoxides include zirconium tetramethoxide, zirconium tetraethoxide, zirconium tetrapropoxide, zirconium tetraisopropoxide, zirconium tetrabutoxide and the like, of which zirconium tetraethoxide is preferred. Examples of the tin tetraalkoxides include tin tetramethoxide, tin tetraethoxide, tin tetrapropoxide, tin tetraisopropoxide, tin tetrabutoxide, tin tetra-t-butoxide and the like, of which tin tetra-t-butoxide is preferred. Examples of 35 aluminum trioxides include aluminum (III) ethoxide, aluminum (III) isopropoxide, aluminum (III) S-butoxide, aluminum (III) n-butoxide, mono-S-butoxyaluminum diisopropoxide and the like, of which aluminum (III) S-butoxide, aluminum (III) n-butoxide and mono-S-butoxyaluminum diisopropoxide are preferred. These compounds may be used singly or in combination.

40 The (B) component should preferably be used in amounts of 5 to 500 parts by weight, more preferably from 10 to 200 parts by weight, per 100 parts by weight of the (A) component. If the amount is less than 5 parts by weight, there is some possibility that the resultant film is not always satisfactory with respect to the strength and adhesion. Over 500 parts by weight, the film may become too hard to attain satisfactory adhesion.

45 In order to further improve the adhesion of the primer composition, it is preferred to add, as (C) component, an organopolysiloxane resin of the following average compositional formula (2) to the primer composition



wherein each  $\text{R}^3$  independently represents an unsubstituted or substituted monovalent hydrocarbon group having, preferably from 1 to 10 carbon atoms, and  $c$  is a positive value of from 0.4 to 1.8.

55 Specific examples of the monovalent hydrocarbon group include alkyl groups such as methyl, ethyl, propyl and the like, alkenyl groups such as vinyl allyl and the like, aryl groups such as phenyl, tolyl and the like, cycloalkyl groups such as cyclohexyl, aralkyl groups such as benzyl, phenylethyl and the like, and those groups mentioned above wherein part or all of the hydrogen atoms in the groups are substituted with a halogen atom or a cyano group, e.g. chloromethyl,

trifluoropropyl, cyanoethyl and the like. Of these, the alkyl and aryl groups are preferred. More preferably, methyl or phenyl is mentioned.

If c is a value smaller than 0.4, the resultant organopolysiloxane becomes too hard and satisfactory adhesion will not develop. On the other hand, when c exceeds 1.8, the resultant product is, for example, diorganopolysiloxane such as silicone oil. This is not satisfactory with respect to adhesion.

The organopolysiloxane resin of (C) component can be readily obtained by co-hydrolysis or hydrolysis of chlorosilanes or alkoxy silanes. Preferably, co-hydrolyzates of chlorosilanes or alkoxy silanes of the formulas,  $R_3^2SiX_2$  and  $R_3SiX_3$  wherein  $R_3$  is same as defined above, and X is chlorine or  $OR^2$  in which  $R^2$  is same as defined before, or co-hydrolyzates of chlorosilanes or alkoxy silanes of the formulas,  $R_3^2SiX$  and  $SiX_4$  wherein  $R_3$  and X are, respectively, as defined above, are used. In the above formulas, the group represented by  $R^3$  is preferably methyl or phenyl. When the chlorosilanes or alkoxy silanes of the formulas,  $R_3^2SiX_2$  and  $R_3SiX_3$ , are co-hydrolyzed, the resultant organopolysiloxane resin consists of  $R_3^2SiO_{2/2}$  units (D units) and  $R_3^2SiO_{3/2}$  units (T units). With the co-hydrolysis of the compounds of the formulas,  $R_3^2SiX$  and  $SiX_4$ , the organopolysiloxane resin obtained consists of  $R_3^2Si_{1/2}$  units (M units) and  $SiO_{4/2}$  units (Q units). The ratio between the D and T units and the ratio between the M and Q units are, respectively, so selected that c is within a range of 0.4 to 1.8.

The organopolysiloxane resin usually has a hydroxyl group at both ends thereof.

The amount of the organopolysiloxane should preferably be in the range of from 10 to 900 parts by weight, more preferably from 30 to 300 parts by weight, per 100 parts by weight of the (A) component.

The primer composition of the invention should preferably be dissolved in organic solvents in order to allow easy coating and working operations. The organic solvents used for this purpose are not critical so far as they are able to dissolve the components of the primer composition therein and are volatile in nature. Examples of such solvents include benzene, toluene, xylene, methanol, ethanol, isopropyl alcohol, butanol, ethylene glycol monomethyl alcohol, acetone, methyl ethyl ketone, ligroin, ethyl acetate, hexane, dioxane, tetrahydrofuran and the like. These solvents may be used on their own or in admixture.

The concentration of the primer composition in an organic solvent should preferably be within a range of 1 to 50 wt%. If the concentration is less than 1 wt%, adhesion may become unsatisfactory. On the other hand, when the concentration exceeds 50 wt%, coating and working operations may not proceed smoothly.

The primer composition of the invention is readily obtained by uniformly mixing the components (A) and (B) along with the component (C), if necessary.

Moreover, pigments, heat resistance improvers and other additives may be added within ranges of amounts not impeding the purposes of the invention.

The primer composition of the invention may be coated on adherends made, for example, of metals or synthetic resins by brushing or spraying, followed by air-drying for a time of five minutes to 24 hours, preferably from 10 minutes to 12 hours and adhesion to silicone rubber. Alternatively, after the air-drying, the coated composition may be heated to make a harder film, followed by adhesion to silicone rubber.

The primer composition of the invention can strongly bond together an adherend, on which an acrylic resin or fluorine resin has been electropainted, and a room temperature curable silicone rubber body together. Accordingly, the composition is very useful as a primer for silicone sealants used for buildings and electropainted building materials.

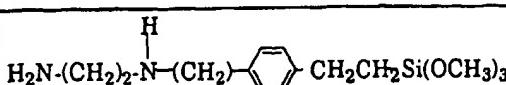
The present invention is more particularly described by way of examples, which should not be construed as limiting the invention. In the examples, parts are by weight. Comparative examples are also described.

#### Examples 1 to 9 and Comparative Examples 1 to 6

Different primer compositions were prepared by uniformly mixing components at mixing ratios indicated in Tables 1 and 2.

An aluminum plate on which an acrylic resin paint AG 300 (commercial name of Kansai Paint Co., Ltd.) had been electropainted and an aluminum plate on which a fluorine resin paint Dick Flow S (commercial name of Kansai Paint Co., Ltd.) were, respectively, provided as an adherend. The surfaces of these plates were each washed with toluene and applied with each primer composition by brushing, followed by air-drying to form a primer film. Subsequently, a room temperature curable silicone rubber of the condensation type (Sealant New 70 of Shin-Etsu Chemical Co., Ltd.) was applied onto the primer film surface and cured. After 14 days, the adhesion between the cured silicone rubber and each of the painted aluminum plate was determined according to the method described in JIS-A-5758. The results are shown in Tables 3 and 4.

Table 1

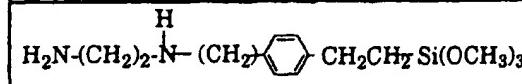
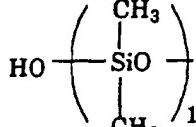
		Example								
		1	2	3	4	5	6	7	8	9
5		2.5	2.5	2.5	2.5	2.5	2.5	0	0	0
										
10	<chem>H2N-CH2-C6H4-CH2CH2-Si(OCH3)3</chem>	0	0	0	0	0	0	2.5	2.5	2.5
	* methylphenylpolysiloxane resin	2.5	2.5	2.5	5.0	2.5	2.5	2.5	5.0	1.5
	titanium tetraethoxide	0	2.5	0	2.5	2.5	1.0	2.5	7.5	0
	zirconium tetrakisopropoxide	2.5	0	2.5	0	0	0	0	0	2.5
	hexane	100	0	50	100	100	100	100	50	100
15	toluene	0	100	50	0	0	0	0	50	0

\* Co-hydrolyzate of methyltrichlorosilane and methylphenyldichlorosilane

(corresponding to a resin of the afore-indicated formula (2) wherein c = 0.9).

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Table 2

		Comparative Example					
		1	2	3	4	5	6
25		2.5	0	0	0	0	2.5
							
30	<chem>H2N-CH2-C6H4-CH2CH2-Si(OCH3)3</chem>	0	0	0	0	0	0
	<chem>H2N-(CH2)2-Si(OCH2CH3)3</chem>	0	0	2.5	0	2.5	0
	<chem>HS-(CH2)3-Si(OCH3)3</chem>	0	0	0	2.5	2.5	0
35	methylphenylpolysiloxane resin	2.5	2.5	2.5	2.5	2.5	0
		0	0	0	0	0	2.5
40	titanium tetraethoxide	0	2.5	2.5	2.5	2.5	2.5
	zirconium tetrakisopropoxide	0	0	0	0	0	0
	hexane	100	0	0	0	0	0
45	toluene	0	100	100	100	100	100

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Table 3

Adherend		Example								
		1	2	3	4	5	6	7	8	9
Acrylic resin paint electropainted aluminum plate	rubber breakage (%)	100	100	100	100	100	100	100	100	100
	interfacial breakage (%)	0	0	0	0	0	0	0	0	0
Fluorine resin paint electropainted aluminum plate	rubber breakage (%)	100	100	100	100	100	100	100	100	100
	interfacial breakage (%)	0	0	0	0	0	0	0	0	0

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Table 4

Adherend		Comparative Example					
		1	2	3	4	5	6
Acrylic resin paint electropainted aluminum plate	rubber breakage (%)	0	0	0	0	35	15
	interfacial breakage (%)	100	100	100	100	65	85
Fluorine resin paint electropainted aluminum plate	rubber breakage (%)	0	0	0	0	30	10
	interfacial breakage (%)	100	100	100	100	70	90

Examples 10 to 16 and Comparative Examples 7 to 13

Different primer compositions were further prepared by uniformly mixing components at mixing ratios indicated in Tables 5 and 6.

Aluminum plates on which acrylic resin paints AG 300 and AG 200 (commercial names of Kansai Paint Co., Ltd.) had been, respectively, electropainted, and an aluminum plate on which a fluorine resin paint Dick Flow S (commercial name of Kansai Paint Co., Ltd.) were, respectively, provided as an adherend. The surfaces of these plates were each washed with toluene and applied with each primer composition by brushing, followed by air-drying to form a primer film. Subsequently, a room temperature curable silicone rubber of the condensation type (Sealant New 70 of Shin-Etsu Chemical Co., Ltd.) was applied onto the primer film surface and cured. The adhesion between the cured silicone rubber and each of the painted aluminum plate after 14 days was determined as an initial adhesion. Moreover, each sample was subjected to an immersion test for 7 days wherein the sample was immersed in water at 50°C. Thereafter, the adhesion of the immersed sample was determined as an adhesion after immersion in water. These adhesions were determined by the method described in JIS-A-5758. The results are shown in Tables 7 and 8.

Table 5

	Example						
	10	11	12	13	14	15	16
5	H <chem>H2N-(CH2)2-N-(CH2)-c1ccccc1-(CH2)2-Si(OCH3)3</chem>	2.5	0	2.5	2.5	2.5	2.5
10	<chem>H2N-CH2-c1ccccc1-CH2CH2-Si(OCH3)3</chem>	0	2.5	0	0	0	0
15	titanium tetraethoxide	2.5	2.5	0	0	0	2
20	zirconium tetraisopropoxide	0	0	2.5	0	0	0.5
25	tin tetra-t-butoxide	0	0	0	2.5	0	0.5
30	aluminum tri-s-butoxide	0	0	0	0	2.5	0
35	hexane	90	90	90	90	90	90
40	isopropanol	10	10	10	10	10	10

Table 6

	Comparative Example						
	7	8	9	10	11	12	13
25	H <chem>H2N-(CH2)2-N-(CH2)-c1ccccc1-(CH2)2-Si(OCH3)3</chem>	2.5	0	0	0	0	0
30	<chem>H2N-CH2-c1ccccc1-CH2CH2-Si(OCH3)3</chem>	0	0	0	0	0	0
35	<chem>H2NC3H6Si(OC2H5)3</chem>	0	0	0	0	0	2.5
40	<chem>H2NC2H4NHC3H6Si(OCH3)3</chem>	0	0	0	0	0	2.5
45	titanium tetraethoxide	0	2.5	0	0	0	2.5
50	zirconium tetraisopropoxide	0	0	2.5	0	0	0
55	tin tetra-t-butoxide	0	0	0	2.5	0	0
60	aluminum tri-s-butoxide	0	0	0	0	2.5	0
65	hexane	90	90	90	90	90	90
70	isopropanol	10	10	10	10	10	10

Table 7

	Adherend	Example						
		10	11	12	13	14	15	16
45	Acrylic resin paint AG 300-electropainted aluminum plate	initial rubber breakage (%)	100	100	100	100	100	100
50		initial interfacial breakage (%)	0	0	0	0	0	0
55		rubber breakage after immersion in water (%)	100	100	100	100	100	100
60		interfacial breakage after immersion in water (%)	0	0	0	0	0	0

Table 7 (continued)

	Adherend	Example						
		10	11	12	13	14	15	16
5	Acrylic resin paint AG 200-electropainted aluminum plate	initial rubber breakage (%)	100	100	100	100	100	100
		initial interfacial breakage (%)	0	0	0	0	0	0
		rubber breakage after immersion in water (%)	100	100	100	100	100	100
		interfacial breakage after immersion in water (%)	0	0	0	0	0	0
10	Fluorine resin paint-electropainted aluminum plate	initial rubber breakage (%)	100	100	100	100	100	100
		initial interfacial breakage (%)	0	0	0	0	0	0
		rubber breakage after immersion in water (%)	100	100	100	100	100	100
		interfacial breakage after immersion in water (%)	0	0	0	0	0	0
15		initial rubber breakage (%)	100	100	100	100	100	100
		initial interfacial breakage (%)	0	0	0	0	0	0
		rubber breakage after immersion in water (%)	100	100	100	100	100	100
		interfacial breakage after immersion in water (%)	0	0	0	0	0	0
20		initial rubber breakage (%)	100	100	100	100	100	100
		initial interfacial breakage (%)	0	0	0	0	0	0
		rubber breakage after immersion in water (%)	100	100	100	100	100	100
		interfacial breakage after immersion in water (%)	0	0	0	0	0	0

Table 8

	Adherend	Comparative Example						
		7	8	9	10	11	12	13
25	Acrylic resin paint AG 300-electropainted aluminum plate	initial rubber breakage (%)	0	0	0	0	0	100
		initial interfacial breakage (%)	100	100	100	100	100	0
		rubber breakage after immersion in water (%)	0	0	0	0	0	0
		interfacial breakage after immersion in water (%)	100	100	100	100	100	100
30	Acrylic resin paint AG 200-electropainted aluminum plate	initial rubber breakage (%)	0	0	0	0	0	100
		initial interfacial breakage (%)	100	100	100	100	100	0
		rubber breakage after immersion in water (%)	0	0	0	0	0	0
		interfacial breakage after immersion in water (%)	100	100	100	100	100	100
35		initial rubber breakage (%)	0	0	0	0	0	100
		initial interfacial breakage (%)	100	100	100	100	100	0
		rubber breakage after immersion in water (%)	0	0	0	0	0	0
		interfacial breakage after immersion in water (%)	100	100	100	100	100	100
40	Fluorine resin paint-electropainted aluminum plate	initial rubber breakage (%)	0	0	0	0	0	100
		initial interfacial breakage (%)	100	100	100	100	100	0
		rubber breakage after immersion in water (%)	0	0	0	0	0	0
		interfacial breakage after immersion in water (%)	100	100	100	100	100	100
45		initial rubber breakage (%)	0	0	0	0	0	100
		initial interfacial breakage (%)	100	100	100	100	100	0
		rubber breakage after immersion in water (%)	100	0	0	0	0	100
		interfacial breakage after immersion in water (%)	0	100	100	100	0	0

Examples 17 to 23 and Comparative Examples 14 to 20

Different primer compositions were prepared by uniformly mixing components at mixing ratios indicated in Tables 9 and 10.

Aluminum plates on which acrylic resin paints AG 300 and AG 200 (commercial names of Kansai Paint Co., Ltd.) had been, respectively, electropainted, and an aluminum plate on which a fluorine resin paint Dick Flow S (commercial name of Kansai Paint Co., Ltd.) were, respectively, provided as an adherend. The surfaces of these plates were each washed with toluene and applied with each primer composition by brushing, followed by air-drying to form a primer

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film. Subsequently, a room temperature curable silicone rubber of the condensation type (Sealant New 70 of Shin-Etsu Chemical Co., Ltd.) was applied onto the primer film surface and cured. The adhesion between the cured silicone rubber and each of the painted aluminum plate after 14 days was determined as an initial adhesion. Moreover, each sample was subjected to an immersion test for 7 days wherein the sample was immersed in water at 50°C. Thereafter, the adhesion of the immersed sample was determined as an adhesion after immersion in water. These adhesions were determined by the method described in JIS-A-5758. The results are shown in Tables 11 and 12.

Table 9

	Example						
	17	18	19	20	21	22	23
H $\text{H}_2\text{N}-\text{CH}_2-\overset{\text{H}}{\underset{ }{\text{N}}}- (\text{CH}_2)_{24}-\text{Si}(\text{OCH}_3)_3$	2.5	0	2.5	2.5	2.5	2.5	2.5
$\text{H}_2\text{N}- (\text{CH}_2)_2-\text{C}_6\text{H}_{10}-\text{Si}(\text{OCH}_3)_3$	0	2.5	0	0	0	0	0
titanium tetraethoxide	2.5	2.5	0	0	0	2	2
zirconium tetrakisopropoxide	0	0	2.5	0	0	0.5	0
tin tetra-t-butoxide	0	0	0	2.5	0	0	0.5
aluminum tri-s-butoxide	0	0	0	0	2.5	0	0
hexane	90	90	90	90	90	90	90
isopropanol	10	10	10	10	10	10	10

Table 10

	Comparative Example						
	14	15	16	17	18	19	20
H $\text{H}_2\text{N}-\text{CH}_2-\overset{\text{H}}{\underset{ }{\text{N}}}- (\text{CH}_2)_{24}-\text{Si}(\text{OCH}_3)_3$	2.5	0	0	0	0	0	0
$\text{H}_2\text{N}- (\text{CH}_2)_2-\text{C}_6\text{H}_{10}-\text{Si}(\text{OCH}_3)_3$	0	0	0	0	0	0	0
$\text{H}_2\text{NC}_3\text{H}_6\text{Si}(\text{OC}_2\text{H}_6)_3$	0	0	0	0	0	2.5	0
$\text{H}_2\text{NC}_2\text{H}_4\text{NHC}_3\text{H}_6\text{Si}(\text{OCH}_3)_3$	0	0	0	0	0	0	2.5
titanium tetraethoxide	0	2.5	0	0	0	2.5	2.5
zirconium tetrakisopropoxide	0	0	2.5	0	0	0	0
tin tetra-t-butoxide	0	0	0	2.5	0	0	0
aluminum tri-s-butoxide	0	0	0	0	2.5	0	0
hexane	90	90	90	90	90	90	90
isopropanol	10	10	10	10	10	10	10

Table 11

	Adherend	Example						
		17	18	19	20	21	22	23
Acrylic resin paint AG 300-electropainted aluminum plate	initial rubber breakage (%)	100	100	100	100	100	100	100
	initial interfacial breakage (%)	0	0	0	0	0	0	0
	rubber breakage after immersion in water (%)	100	100	100	100	100	100	100
	interfacial breakage after immersion in water (%)	0	0	0	0	0	0	0

Table 11 (continued)

		Example						
Adherend		17	18	19	20	21	22	23
5 Acrylic resin paint AG 200-electropainted aluminum plate	initial rubber breakage (%)	100	100	100	100	100	100	100
	initial interfacial breakage (%)	0	0	0	0	0	0	0
	rubber breakage after immersion in water (%)	100	100	100	100	100	100	100
	interfacial breakage after immersion in water (%)	0	0	0	0	0	0	0
10 Fluorine resin paint-electropainted aluminum plate	initial rubber breakage (%)	100	100	100	100	100	100	100
	initial interfacial breakage (%)	0	0	0	0	0	0	0
	rubber breakage after immersion in water (%)	100	100	100	100	100	100	100
	interfacial breakage after immersion in water (%)	0	0	0	0	0	0	0
20								

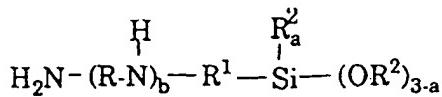
Table 12

		Comparative Example						
Adherend		14	15	16	17	18	19	20
25 Acrylic resin paint AG 300-electropainted aluminum plate	initial rubber breakage (%)	0	0	0	0	0	100	100
	initial interfacial breakage (%)	100	100	100	100	100	0	0
	rubber breakage after immersion in water (%)	0	0	0	0	0	0	0
	interfacial breakage after immersion in water (%)	100	100	100	100	100	100	100
30 Acrylic resin paint AG 200-electropainted aluminum plate	initial rubber breakage (%)	0	0	0	0	0	100	100
	initial interfacial breakage (%)	100	100	100	100	100	0	0
	rubber breakage after immersion in water (%)	0	0	0	0	0	0	0
	interfacial breakage after immersion in water (%)	100	100	100	100	100	100	100
35 Acrylic resin paint AG 200-electropainted aluminum plate	initial rubber breakage (%)	0	0	0	0	0	100	100
	initial interfacial breakage (%)	100	100	100	100	100	0	0
	rubber breakage after immersion in water (%)	0	0	0	0	0	0	0
	interfacial breakage after immersion in water (%)	100	100	100	100	100	100	100
40 Fluorine resin paint-electropainted aluminum plate	initial rubber breakage (%)	0	0	0	0	0	100	100
	initial interfacial breakage (%)	100	100	100	100	100	0	0
	rubber breakage after immersion in water (%)	0	0	0	0	0	0	0
	interfacial breakage after immersion in water (%)	100	100	100	100	100	100	100
45 Fluorine resin paint-electropainted aluminum plate	initial rubber breakage (%)	0	0	0	0	0	100	100
	initial interfacial breakage (%)	100	100	100	100	100	0	0
	rubber breakage after immersion in water (%)	100	0	0	0	0	100	100
	interfacial breakage after immersion in water (%)	0	100	100	100	100	0	0
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**Claims**

1. A primer composition which comprises:

55 (A) an organosilicon compound of the general formula (1)

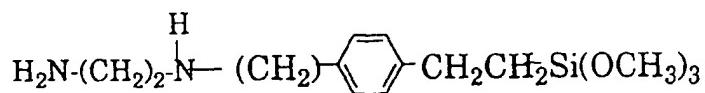


wherein R is C<sub>1</sub>-C<sub>6</sub> alkylene, R<sup>1</sup> is a divalent hydrocarbon group having at least 4 carbon atoms, each R<sup>2</sup>, which may be the same or different, is an unsubstituted or substituted monovalent hydrocarbon group, a is 0 or 1 and b is 0, 1, 2 or 3; and

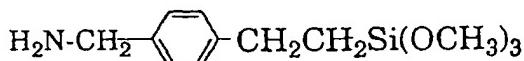
(B) a condensation catalyst for the organosilicon compound (A).

2. A composition according to Claim 1, wherein R is -CH<sub>2</sub>CH<sub>2</sub>-.
- 15 3. A composition according to Claim 1 or 2 wherein R<sup>1</sup> is phenylene or a phenylene-containing divalent hydrocarbon group.
4. A composition according to any one of the preceding Claims wherein each R<sup>2</sup>, which may be the same or different, is C<sub>1</sub>-C<sub>4</sub> alkyl.
- 20 5. A composition according to any one of the preceding Claims wherein a is 0 and b is 0 or 1.
6. A composition according to any one of the preceding Claims wherein the organosilicon compound is a compound of one of the following formulae

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and



7. A composition according to any one of the preceding Claims wherein the catalyst (B) is selected from titanium tetraalkoxides of the formula Ti(OR<sup>4</sup>)<sub>4</sub> and partial hydrolyzates thereof, zirconium tetraalkoxides of the formula Zr(OR<sup>4</sup>)<sub>4</sub> and partial hydrolyzates thereof, tin tetraalkoxides of the formula Sn(OR<sup>4</sup>)<sub>4</sub> and partial hydrolyzates thereof, and aluminum alkoxides Al(OR<sup>4</sup>)<sub>4</sub> and partial hydrolyzates thereof, wherein each R<sup>4</sup> is as defined for R<sup>2</sup> in claim 1.
- 40 8. A composition according to any one of the preceding Claims wherein the catalyst (B) is present in an amount of from 5 to 500 parts by weight per 100 parts by weight of the organosilicon compound (A).
9. A composition according to any one of the preceding Claims which further comprises:

(C) an organopolysiloxane resin of the following average compositional formula

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wherein each R<sup>3</sup>, which may be the same or different, is an unsubstituted or substituted monovalent hydrocarbon group, and c is a number of from 0.4 to 1.8.

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10. A composition according to Claim 9 wherein the organopolysiloxane (C) is present in an amount of 10 to 900 parts by weight per 100 parts by weight of the organosilicon compound (A).

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11. A primer solution which comprises a composition as defined in any one of the preceding Claims dissolved in a solvent therefor at a concentration of 1 to 50 wt%.

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European Patent  
Office

## EUROPEAN SEARCH REPORT

Application Number  
EP 96 30 5575

DOCUMENTS CONSIDERED TO BE RELEVANT			CLASSIFICATION OF THE APPLICATION (Int.Cl.6)						
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim							
X	US-A-4 769 308 (HIRUMA ET AL) * claim 1 *	1-8	C08K5/54 C09D4/00 C09D183/04						
X	US-A-5 300 611 (FUJIOKA ET AL ) * claims 1,19 *	1-8							
X	EP-A-0 220 733 (TORAY)  * page 4, line 18 - page 5, line 23 * * page 6 * * page 7, line 7 - line 35 *	1-5,7,8, 11							
Y	EP-A-0 373 562 (DOW CORNING)  * column 2, line 3 - line 46 *	9,10							
X	EP-A-0 384 401 (DOW TORAY) * page 3, line 9 - line 22; claim 1 *	1-5,7,8, 11							
	-----	9,10							
	-----	-----	TECHNICAL FIELDS SEARCHED (Int.Cl.6)						
			C08K C09D						
<p>The present search report has been drawn up for all claims</p> <table border="1" style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 33%;">Place of search</td> <td style="width: 33%;">Date of completion of the search</td> <td style="width: 34%;">Examiner</td> </tr> <tr> <td>THE HAGUE</td> <td>25 November 1996</td> <td>Lentz, J</td> </tr> </table>				Place of search	Date of completion of the search	Examiner	THE HAGUE	25 November 1996	Lentz, J
Place of search	Date of completion of the search	Examiner							
THE HAGUE	25 November 1996	Lentz, J							
CATEGORY OF CITED DOCUMENTS		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document							
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document									